

**PCT**WORLD INTELLECTUAL PROPERTY ORGANIZATION  
International Bureau

## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<b>(51) International Patent Classification <sup>5</sup> :</b> <b>C10M 137/06, 137/08</b>	<b>A1</b>	<b>(11) International Publication Number:</b> <b>WO 92/07924</b> <b>(43) International Publication Date:</b> 14 May 1992 (14.05.92)
<b>(21) International Application Number:</b> PCT/US91/08087 <b>(22) International Filing Date:</b> 1 November 1991 (01.11.91)  <b>(30) Priority data:</b> 612,400 2 November 1990 (02.11.90) US  <b>(71)(72) Applicant and Inventor:</b> SMITH, Noel, S. [US/US]; 4192 Allegheny, Troy, MI 48098 (US).  <b>(74) Agents:</b> WEINTRAUB, Arnold, S. et al.; 3001 W. Big Beaver Road, Suite 504, Troy, MI 48084 (US).  <b>(81) Designated States:</b> AT (European patent), AU, BE (European patent), BR, CA, CH (European patent), DE (European patent), DK (European patent), ES (European patent), FR (European patent), GB (European patent), GR (European patent), IT (European patent), JP, KR, LU (European patent), NL (European patent), SE (European patent).		<b>Published</b> <i>With international search report.</i>
<b>(54) Title:</b> COMPOSITION AND METHOD FOR TREATING METAL  <b>(57) Abstract</b>  A method and composition for treating a metal workpiece, to reduce galling thereof in a mold. The method involves coating the workpiece with the composition. The coating composition includes: a) a phosphate ester lubricant; b) an acrylic polymer; c) a polyolefin; and d) water or other suitable solvent. The workpiece may be plated with a metallic compound, such as, tin, zinc phosphate, etc. before the coating is applied thereto.		

**FOR THE PURPOSES OF INFORMATION ONLY**

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AT	Austria	ES	Spain	MG	Madagascar
AU	Australia	FI	Finland	ML	Mali
BB	Barbados	FR	France	MN	Mongolia
BE	Belgium	GA	Gabon	MR	Mauritania
BF	Burkina Faso	GB	United Kingdom	MW	Malawi
BG	Bulgaria	GN	Guinea	NL	Netherlands
BJ	Benin	GR	Greece	NO	Norway
BR	Brazil	HU	Hungary	PL	Poland
CA	Canada	IT	Italy	RO	Romania
CF	Central African Republic	JP	Japan	SD	Sudan
CG	Congo	KP	Democratic People's Republic of Korea	SE	Sweden
CH	Switzerland			SN	Senegal
CI	Côte d'Ivoire	KR	Republic of Korea	SU+	Soviet Union
CM	Cameroon	LI	Liechtenstein	TD	Chad
CS	Czechoslovakia	LK	Sri Lanka	TG	Togo
DE*	Germany	LU	Luxembourg	US	United States of America
DK	Denmark	MC	Monaco		

+ Any designation of "SU" has effect in the Russian Federation. It is not yet known whether any such designation has effect in other States of the former Soviet Union.

COMPOSITION AND METHOD FOR TREATING METAL  
BACKGROUND OF THE INVENTION

Field of the Invention:

The present invention relates to a method of and composition for treating a metal workpiece to prevent or reduce galling of the workpiece in a mold or die. More particularly, the present invention relates to such a method and composition in which a coating thereof is applied to the outside of the workpiece to prevent or reduce galling thereof in a high pressure forming operation.

Prior Art:

It is well known in the metal forming art, that a metal slug may be placed into a mold having a desired shape. A rod or similar device is then rammed down into the slug with high force, causing the metal to deform to the contours of the mold. Unfortunately, in this process, friction is often created between the metal slug and the wall of the mold, causing galling or a non-smooth exterior surface of the finished molded part.

To minimize this condition lubricant compositions have been developed. However, presently known lubricants do not withstand use in high pressure applications. Furthermore, such prior art lubricants often times do not evidence requisite stability when applied to a metal workpiece.

Thus, there is a need in the metal forming art for a lubricant coating which is stable after application to a metal workpiece, and which minimizes galling of the workpiece during a high pressure forming operation.

SUMMARY OF THE INVENTION

The present invention provides a method of and composition for treating a metal workpiece, such as a slug or plate, to reduce galling thereof in a mold or die. The

present, composition an aqueous lubricant composition comprising:

- (a) film forming compound capable of forming a film on a metallic surface;
- (b) a polyolefin;
- (c) a coalescent for promoting film formation on the metal surface; and
- (d) water.

Conventionally, the aqueous composition is prepared from a lubricant concentrate comprising:

- (a) a film forming compound capable of forming a film on a metallic surface;
- (b) polyolefin;
- (c) a coalescent; and,
- (d) water.

Alternatively, a suitable powder from which the aqueous lubricant composition and concentrate may be provided. Such powder, generally, comprises:

- (a) film-forming compound capable of forming a film on the surface of a metal;
- (b) a polyolefin; and
- (c) a coalescent.

The powder is then added to water to form the composition. The composition may further include pigments, high pressure additives, solubilizers, additional lubricants and the like. The composition, ordinarily, contains from about 5% to about 25%, by weight of solids based on the total weight. Usually, the acrylic polymer and the polyolefin are present in a 0.25:1 to 2:1 respective weight ratio.

The concentrate, preferably, comprises from about 5 percent to about 80 percent, by weight, of solids. Likewise, the film forming compound and polyolefin are present in a respective weight ratio from about 0.25:1 to about 2:1. The use solution is prepared by diluting the

concentrate with water at ambient conditions with stirring.

In use in the lubricant coating is applied directly to a metal part by immersion, spraying or the like at ambient temperatures or higher, up to about 150°F, and at ambient pressure.

The metal part to which the coating is applied may be pre-treated, such as by plating, heat treating, or phosphating, etc. prior to the lubricant coating being applied. However, the present invention will work with metal parts that are not pre-treated of the metal and/or the forming operation.

For a more complete understanding of the present invention, reference is made to the following detailed description, and the examples contained therein.

#### DESCRIPTION OF THE PREFERRED EMBODIMENTS

It has been discovered, in accordance with the practice of the present invention, that a unique combination of compounds creates a composition which significantly improves molding performance properties when used as a coating on a metal workpiece to prevent galling thereof in a mold in an extreme pressure stamping or forming operation.

The composition of the present invention may be applied as a water-based coating to the exterior of a steel slug or other metal workpiece before the workpiece is placed in a mold or die. The lubricating coating composition is then allowed to dry at room temperature or forced air dried.

The composition of the present invention, generally, comprises:

- (a) a film-forming composition;
- (b) a polyolefin;
- (c) a coalescent; and
- (d) water.

The coating composition hereof, generally,

comprises from about 1 percent to about 30 percent, by weight of solids, based on the total weight, and, preferably, from about 5 percent to about 25 percent, by weight, based on the total weight.

The film-forming composition and polyolefin are present in a respective weight ratio of from about 0.25:1 to about 2:1 and, preferably about 0.75:1 to about 1.25:1.

The lubricant-coating composition, is, preferably, prepared from a concentrate comprising:

- (a) a film-forming composition;
- (b) a polyolefin;
- (c) a coalescent; and
- (d) water.

The concentrate, generally, comprises from 5 percent to about 80 percent, by weight, of solids in the weight ratios denoted above. Preferably, the concentrate comprises from about 40 percent to about 60 percent by weight of the solids.

The use solution is prepared by diluting the concentrate with water. Of course, the use solution may be prepared directly from the requisite ingredients.

The film-forming component hereof is one which is capable of forming a film on the surface of a metal. Suitable film-forming compounds include, for example, acrylic polymers, styrene-acrylic copolymers, and other thermoplastic compounds. Preferably, the film-forming compound is an acrylic polymer, a styrene-acrylic copolymer or mixtures thereof.

The acrylic polymer, where used, is, preferably, water-soluble and acts as a barrier and as a film former on a part to be formed. As a barrier, the polymer functions to prevent direct contact between the tool or mold or die and the workpiece. As a film former, the acrylic polymer forms a dry film matrix for promoting polyolefin adhesion and distribution. Furthermore, the acrylic promotes the

dispersion of the polyolefin in the aqueous coating composition.

Acrylic polymers which are suitable for use in the practice of the present invention are preferably polyacrylates and the copolymers of styrene or styrene derivatives with acrylic acid or derivatives thereof and include solid polymers in a dispersed form and mixtures of polymers with other materials.

Typically, and is known to those skilled in the art, these polymers are prepared by the copolymerization of (1) styrene and/or alpha methyl styrene and (2) acrylic acid, methacrylic acid, 2-ethyl hexyl acrylate, methyl methacrylate, butyl acrylate and the like, as well as mixtures thereof. Usually the styrene component comprises from about 0 percent to about 80 percent by weight of the polymer and the acrylate component from about 40 percent by weight of the polymer.

In the practice of the present invention the polymer has a molecular weight of at least 2000 and, generally, from about 5000 to about 250,000 and an acid number of less than 250.

One particularly useful, commercially available product, which is suitable for use in the practice of the present invention is a mixture of styrene-acrylic polymer with one to three percent diethylene glycol monoethyl ether sold by S.C. Johnson & Son, Incorporated in Racine, Wisconsin under the name JONCRYL 678 acrylic resin. This product is described as a solid, clear flakes having an acid value of 2000; a density of 1.25g/cc, a molecular weight of 8,000.

Another useful product is sold commercially under the name JONCRYL 537 which is a 2000,00 molecular weight styrene-acrylic acid copolymer having the following physical properties;

Non-volatiles	46%	Filming Temperature	42°C
pH	9	Glass transition temp.	44°C
Brookfield		Density g/cc	1.05
Viscosity, cps	150		
Acid No.	40		

Likewise, mixtures of these two compound can be used.

It should be noted that although thermoplastic film formers are preferred, it is possible to use thermoset components such as, for example, epoxy resins and the like.

The film forming compound generally comprises from about 2.5 percent to about 35 percent, by weight, based on the total weight of the concentrate, and preferably from about 15 to about 30 percent by weight, based on the total weight of the concentrate.

The polyolefin provides a means for reducing the coefficient of friction between the tool and the workpiece. The polyolefin is compatible with the film-forming compound and is readily suspended thereby in the aqueous medium. Useful polyolefins include polyethylene, polypropylene and copolymers thereof as well as halogenated polyolefins thereof.

Usually, in commercial applications the polyolefin is available as a blend of various polyolefins and halogenated polyolefins. The polyolefin is, ordinarily, present in the concentrate in an amount ranging from about 2.5 percent to about 35 percent by weight, and preferably from about 15 to about 30 percent by weight, of the total weight of the concentrate.

The preferred polyolefin for use in the practice of the present invention is polyolefin blend of polyethylene and halogenated polyethylene, such as, e.g., fluorinated polymeric polyolefins or chlorofluorinated polyolefins. A particularly preferred and commercially available polyolefin is that which is marked by Daniel Product Company of Jersey City, New Jersey, under the



trade name SLIP-AYD 600. This is described as a dry, micronized powder containing 100 percent solids with an average particle size of 1.5m, a maximum particle size of 10.0m, a specific gravity of 1.17 and is a blend of halogenated polyolefins, polyethylenes and other synthetic waxes. Another product which is suitable for use in the practice of the present inventions is a water-soluble dispersion sold by Daniel Products under the trade name SLIP-AYD SL 630 and which contains about 40 percent by weight of the SL 600, 15 percent of acrylic solid, 43.0 percent water, and 2.0 percent isopropanol. The product has a flash point of 120°.

The coalescent serves to promote film formation on the metal surface. Although not wishing to be bound by any theory it appears that in an aqueous system, the coalescent aids in micelle formation by the film forming component in the aqueous phase. Furthermore, when the water is removed, the coalescent makes the micelle tacky so that a film is formed with the olefin being trapped therewithin.

The coalescent contemplated for use herein is one which promotes micelle formation, functions as a surfactant in the aqueous solution and which, preferably, is an extreme pressure additive.

Useful coalescents include, for example, alkanols diols, triols, glycol ether, butylcellosolve, nonionic surfactants, anionics surfactants, cationic surfactants, phosphate ethers and esters, polyesters, polyethers and the like as well as mixtures thereof.

Useful anionic wetting agents include those sold by American Cyanamid under the name AEROSOL OT and include that which is defined in U.S. Patent No. 2,441,341, the disclosure of which is hereby incorporated by reference. These anionic compounds are the alkali metal salts of carboxylic acids and specifically the sodium salt of dioctyl sulfosuccinate acid. A particularly preferred such

compound is that sold under the name AEROSOL OT-75% which is water-soluble and contains 75 percent active ingredient and approximately 5 percent of a lower alcohol.

Other use coalescents include alkoxylated phosphate ester-type lubricants.

The coalescent is present in the use solution in an amount ranging from about 0.1 percent to about 1 percent by weight, based on the total weight of the use solution and, preferably, of concentrate from about 0.25 percent to about 0.85 percent by weight, based on the total weight. Where incorporated into the concentrate, the coalescent is present in an amount ranging from about 0.5 percent to about 10 percent by weight, based on the total weight, and preferably from about 2.5 percent to about 4 percent by weight.

One suitable phosphate ester lubricant for use in the practice of the present invention is a triethanol amine salt of a polyoxyethylene nonionic surfactant, such as those sold commercially by GAF Corporation as ANTARA lubricants. These products are marked as extreme pressure additive lubricants and rust inhibitors. The ANTARA lubricants contain a phosphate radical on a polyoxyethylene-nonionic surfactant base. They are generally translucent yellowish fluids at standard temperature and pressure, with a specific gravity in the range of 1.0-1.2, with a moisture content from 0.5 percent to about 2 percent, and when diluted in water to a 10 percent solution, exhibit a pH from 1.5 percent to about 2.5 percent. Their phosphorous content ranges from 2.8 percent to about 6.0 percent by weight. These compounds are soluble in aromatic solvents and are generally soluble or self-emulsifying in water solution.

In the practice of the present invention, the preferred coalescent is the alkoxylated phosphate-ester. This coalescent not only functions as a coalescent and

surfactant, but is an extreme or higher pressure additive, as well as a plasticizer. This is important in that it prevents adhesion of the polyolefin to the workpiece and/or tool.

Other use adjuvants include defoamers, solubilizers, pigments, biocides and the like.

The defoamer where used is present in the use solution in an amount ranging from about 0.1 percent to about 1 percent by weight, based on the total weight. Any defoamer may be used herein so long as the defoamer selected is non-reactive with the components and does not detract from the lubricating.

To promote the dispersion and solubilizing of the wetting agent and/or the acrylic polymer as well as to neutralize any residual acid groups a solubilizing agent may, also, be incorporated. The solubilizing agent is present in an amount ranging from about 0 percent to about 3 percent by weight, and, preferably, from about 0.5 percent to about 2 percent based on the total weight of the concentrate.

Preferred solubilizing agents are the alkanol amines and alkyl amines, such as, monethanol amine, diithanolamine, triithanolamine, isopropanolamine, diesopropanolamine, trisopropanolamine, monethylamine, butylamine, and the like, as well as mixtures thereof. Also, ammonia may be used as a solubilizer.

As noted pigments may be incorporated where it is desirable to color the metallic workpiece.

The composition hereof, as noted, is used to reduce galling of metal parts, such as sparkplug bases and the like. As noted, the composition is applied to the metal piece to be formed by immersion, spraying or the like. Although not essential it should be noted that the metal workpiece may be plated prior to the application of the present lubricant. The present composition is

applicable to tall types of metals, including, for example, steel, iron, stainless steel, zinc, etc., which may or may not be plated with alumina ( $Al_2O_3$ ), chromium, nickel, zinc phosphate, zinc and tin etc. Where the metal is plated and formed under extreme pressure, zinc phosphate is preferred as a plating agent. Plating of the metal workpiece, in accordance with the present invention, may be accomplished by electroplating or by other methods known to those in the art.

In use, the coating is applied directly to the metal workpiece to be formed by immersion, spraying or the like. Ordinarily, the coating is applied at room temperature or higher, up to about 150° to 160°F. Thereafter, the workpiece is dried to eliminate the water leaving a film on the workpiece.

The composition of the present invention generally dries as a thin, dry, lubricious coating around the metal workpiece. After the coating is dried, the metal workpiece is then placed in a mold and stamped or pressed into the mold or die in a conventional fashion. Alternatively, the workpiece can be stored for extended periods of time such as, e.g., weeks or months, between the coating thereof with the composition of the present invention, and the forming operation.

The use solution usually contains from about 5 percent to about 30 percent by weight of solids, based on the total weight and as a pH range from about 7 percent to 9 percent, and preferably from about 8.5 percent to about 9 percent.

The present coating composition has particularly utilizing in the treatment of steel slugs such as those used in sparkplug manufacture. Typically such steel slugs are processed in accordance herewith by:

- (a) cleaning the annealed slug;
- (b) rinsing the slug;

- (c) phosphating the slug;
- (d) re-rinsing the slug;
- (e) drying the slug;
- (f) coating the slug with the lubricant-coating composition hereof; and,
- (g) drying the slug to form the film thereon.

It should be noted that where a titanium coated punch is employed on the metal forming operation., phosphating of the workpiece can be eliminated.

Likewise in stamping a workpiece, the phosphating can be eliminated.

It should be noted with respect to the present invention that it has been described herein with respect to an aqueous system. As noted hereunder, it is possible to employ a powder of the film-forming compound, preferably, the film forming component; the polyolefin, and the coalescent. This powder can then be admixed with a suitable solvent to form the coating composition.

In this regard, it is to be noted, that although the present invention, in its preferred form, is employed as an aqueous system, the present invention is not to be construed as being so-limited. Rather, suitable solvents may be used in lieu of water, such as, for example, glycol ethers, hexane, heptane, benzene, toluene, and the like. The only criteria to be attached to the solvent is that it not detract from the ability of the film-forming compound to adhere to the surface of the metal. Furthermore, the solvent must be capable of drying at an ambient or slightly elevated temperatures. However, and as noted hereinabove, in the practice of the present invention, it is preferred that an aqueous system be employed.

It should further be noted with respect to the present invention that it has been discovered that the present composition utility as a torque-tension coating composition for plated workpieces which are either zinc,

cadmium, tin, dechioniate plated. In other words, the present composition, when applied to a metal workpiece, which is threadingly secured to another workpiece, reduces the forces necessary to apply torque to the part to be threadingly attached.

Traditionally, metal workpieces, especially in automotive applications, have a cadmium and wax coating applied thereto prior to the application of the torque thereto. The present invention eliminates the need for the cadmium and wax coating while permitting the reduction of the torque with increase in the tension. As a torque-tension composition, the composition is applied, preferably, as an aqueous composition, the composition is applied, preferably, as an aqueous composition dried, leaving the film-forming component having the coalescent and polyolefin present therewithin.

For a more complete understanding of the present invention, reference is made to the following examples. In the examples, which are to be construed as illustrative rather than limitative, all parts are by weight, absent indications to the contrary.

Example 1

This example illustrates the preparation of a coating composition in accordance with the present invention.

A composition is prepared by adding to a suitable vessel, at room temperature, the phosphate ester, acrylic polymer, and polyolefin. Water as a solvent is, then, added to other ingredients. The ingredients were then thoroughly mixed together. The following table sets forth the ingredients and their amounts:

<u>INGREDIENT</u>	<u>AMOUNT, pbw</u>
COALESCENT <sup>1</sup>	0.5
ACRYLIC POLYMER FILM FORMER <sup>2</sup>	4.0
POLYOLEFIN <sup>3</sup>	10.0
WATER, DISTILLED	85.5

---

(1) A commercially available phosphate ester lubricant sold by GAF Corporation under the name "ANTARA LE-700"

(2) A commercially available acrylic resin sold by Johnson Wax under the name "JONCRYL 678"

(3) A commercially available mixture of halogenated polyolefins, polyethylenes, and other synthetic waxes sold by Daniel Products Company under the name "SLIP-AYD SL 600"

A steel slug was coated with tin by conventional

electroplating techniques. The tin plated steel slug was then coated with the composition of example 2 and was allowed to air dry. The slug was then molded in a stamping press, and significantly reduced galling of the exterior surface of the molded part was observed.



Example II

This example illustrates the preparation of a concentrate for use in preparing a lubricating-coating composition in accordance with the present invention.

Into a suitable vessel, with mixing was added the following:

<u>INGREDIENT</u>	<u>AMT. pbw</u>
FILM FORMING COMPOUND <sup>(1)</sup>	24.32
POLYOLEFIN <sup>(2)</sup>	63.22
COALESCENT 1 <sup>(3)</sup>	3.66
COALESCENT 2 <sup>(4)</sup>	0.49
DEFOAMER <sup>(5)</sup>	0.29
TRISOPROPANOLAMINE	3.50
WATER, DISTILLED	4.57

This concentrate which is substantially a 50 percent solid concentration, is then diluted with sufficient water to form a 10 percent solid use solution.

---

(1) a styrene-acid copolymer acrylic sold by S.G. Johnson under the name JONCRYL 537.

(2) a dispersion sold by Daniel Products under the name SLIP-AYD SL 630

(3) a phosphate ester lubricant sold by Mazer chemical under the name MASLIP 504

(4) AEROSOL OT 75 percent anionic wetting agent

(5) an emulsion of paraffin based mineral oils and hydrophobic components sold by BYK Chemie under the name BYK-32

Thereafter, a steel slug was coated with the above composition, by dipping into the solution, and allowing it to air dry. The coated slug was then molded in a press to form a spark plug base. Significantly reduced galling of the exterior surface of the molded part was observed as compared to an uncoated slug.

Having, thus, described the invention, what is claimed is:

CLAIMS

1. A concentrate for preparing lubricant coating composition for a metal workpiece the concentrate comprising:

a) a film forming composition capable of forming a film on the workpiece,

b) a polyolefin, and

c) a coalescent.

2. The concentrate composition of Claim 1 wherein the film forming composition and polyolefin are present in a respective weight ratio of about 0.25:1 to about 2:1.

3. The concentrate composition of Claim 1 which further comprises:

(a) from about 2.5 percent to about 35 percent, by total weight, of the film forming composition,

(b) from about 2.5 percent to about 35 percent, by total weight, of the polyolefin,

(c) from about 0.5 percent to about 10.0 percent by total weight, of the coalescent, and

(d) from about 80 percent to about 94.5 percent, by total weight of water.

4. The concentrate composition of Claim 3 wherein the concentrate is a aqueous concentrate comprising from about 5 percent to about 80 percent, by weight, of solids, based on the total weight.

5. The concentrate composition of Claim 4,

wherein the film forming compound is selected from the group consisting of acrylic and acid polymers, styrene-acrylic acid copolymers and mixtures thereof.

6. The concentrate composition of Claim 4 wherein the polyolefin is selected from the group consisting of polyethylene, polypropylene, halogenated polyethylene, halogenated polypropylene and mixtures thereof.

7. The concentrate composition of Claim 2 wherein the coalescent is a phosphate ester.

8. The concentrate composition of Claim 4 wherein the concentrate is a powder.

9. A lubricant coating use composition for a metal workpiece, comprising:

(a) from about 70 to 95 percent, by weight, of a solvent, and

(b) from about 30 to 5 percent, by weight, of the concentrate of Claim 4, and wherein the use composition comprises from at least five percent solids based on the total weight.

# INTERNATIONAL SEARCH REPORT

International Application No. PCT/US91/08087

<b>I. CLASSIFICATION OF SUBJECT MATTER</b> (if several classification symbols apply, indicate all) <sup>6</sup> According to International Patent Classification (IPC) or to both National Classification and IPC IPC(5): C10M 137/06, C10M 137/08														
<b>II. FIELDS SEARCHED</b> <div style="text-align: right; font-size: small;">Minimum Documentation Searched <sup>7</sup></div> <table style="width: 100%; border: none;"> <tr> <td style="width: 20%; border: none;">Classification System</td> <td style="border: none;">Classification Symbols</td> </tr> <tr> <td style="border: 1px solid black; padding: 5px; vertical-align: top;">US</td> <td style="border: 1px solid black; padding: 5px; vertical-align: top;">252/32.5; 42; 49.3; 49.5; 56R; 565; 58 72/42</td> </tr> </table> <div style="text-align: center; font-size: x-small; margin-top: 5px;">           Documentation Searched other than Minimum Documentation            to the Extent that such Documents are Included in the Fields Searched <sup>8</sup> </div>			Classification System	Classification Symbols	US	252/32.5; 42; 49.3; 49.5; 56R; 565; 58 72/42								
Classification System	Classification Symbols													
US	252/32.5; 42; 49.3; 49.5; 56R; 565; 58 72/42													
<b>III. DOCUMENTS CONSIDERED TO BE RELEVANT <sup>9</sup></b> <table border="1" style="width: 100%; border-collapse: collapse;"> <thead> <tr> <th style="width: 10%; font-size: x-small;">Category <sup>10</sup></th> <th style="width: 70%; font-size: x-small;">Citation of Document, <sup>11</sup> with indication, where appropriate, of the relevant passages <sup>12</sup></th> <th style="width: 20%; font-size: x-small;">Relevant to Claim No. <sup>13</sup></th> </tr> </thead> <tbody> <tr> <td style="text-align: center; vertical-align: top;">Y</td> <td style="vertical-align: top;">US, A, 4,752,405 21 JUNE 1988 See entire document.</td> <td style="text-align: center; vertical-align: top;">ALL</td> </tr> <tr> <td style="text-align: center; vertical-align: top;">Y</td> <td style="vertical-align: top;">US, A, 4,654,155 31 MARCH 1987 See entire document.</td> <td style="text-align: center; vertical-align: top;">ALL</td> </tr> <tr> <td style="text-align: center; vertical-align: top;">Y</td> <td style="vertical-align: top;">US, A, 3,725,274 03 APRIL 1973 See entire document.</td> <td style="text-align: center; vertical-align: top;">ALL</td> </tr> </tbody> </table>			Category <sup>10</sup>	Citation of Document, <sup>11</sup> with indication, where appropriate, of the relevant passages <sup>12</sup>	Relevant to Claim No. <sup>13</sup>	Y	US, A, 4,752,405 21 JUNE 1988 See entire document.	ALL	Y	US, A, 4,654,155 31 MARCH 1987 See entire document.	ALL	Y	US, A, 3,725,274 03 APRIL 1973 See entire document.	ALL
Category <sup>10</sup>	Citation of Document, <sup>11</sup> with indication, where appropriate, of the relevant passages <sup>12</sup>	Relevant to Claim No. <sup>13</sup>												
Y	US, A, 4,752,405 21 JUNE 1988 See entire document.	ALL												
Y	US, A, 4,654,155 31 MARCH 1987 See entire document.	ALL												
Y	US, A, 3,725,274 03 APRIL 1973 See entire document.	ALL												
<div style="display: flex; justify-content: space-between; font-size: x-small;"> <div style="width: 45%;"> <p><sup>10</sup> Special categories of cited documents:</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> </div> <div style="width: 45%;"> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.</p> <p>"A" document member of the same patent family</p> </div> </div>														
<b>IV. CERTIFICATION</b> <table style="width: 100%; border: none;"> <tr> <td style="width: 50%; border: none;">Date of the Actual Completion of the International Search</td> <td style="width: 50%; border: none;">Date of Mailing of this International Search Report</td> </tr> <tr> <td style="border: none; text-align: center;">21 JANUARY 1992</td> <td style="border: none; text-align: center; font-size: large;">27 FEB 1992</td> </tr> <tr> <td style="border: none;">International Searching Authority</td> <td style="border: none;">Signature of Authorized Officer</td> </tr> <tr> <td style="border: none; text-align: center;">ISA/US</td> <td style="border: none; text-align: center;">             MARIA NUZZOLILLO         </td> </tr> </table>			Date of the Actual Completion of the International Search	Date of Mailing of this International Search Report	21 JANUARY 1992	27 FEB 1992	International Searching Authority	Signature of Authorized Officer	ISA/US	 MARIA NUZZOLILLO				
Date of the Actual Completion of the International Search	Date of Mailing of this International Search Report													
21 JANUARY 1992	27 FEB 1992													
International Searching Authority	Signature of Authorized Officer													
ISA/US	 MARIA NUZZOLILLO													

**THIS PAGE BLANK (USPTO)**